

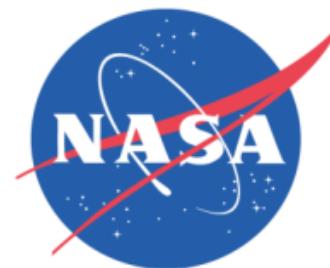


# Joining and Integration of Silicon Carbide-Based Ceramic Materials for Aerospace Applications: An Overview

Michael C. Halbig<sup>1</sup> and Mrityunjay Singh<sup>2</sup>

<sup>1</sup>NASA Glenn Research Center, Cleveland, OH

<sup>2</sup>Ohio Aerospace Institute, Cleveland, OH



**47th International Conference and Exposition on Advanced Ceramics and Composites**  
Daytona Beach, Florida, January 22-27, 2023.

***Emerging Materials & Sustainable Manufacturing Technologies Symposium  
in Honor of Dr. Tatsuki Ohji.***



# Outline

- **Introduction**
  - **Objectives, Benefits, and Applications**
- **NASA GRC Joining Technologies for SiC-based Ceramics: monolithics and CMCs to themselves, each other, and to metals.**
  - **Brazing**
  - **ARCJoinT - Affordable, Robust Ceramic Joining Technology**
  - **Diffusion Bonding**
  - **REABond - Refractory Eutectic Assisted Bonding**
  - **SET Joining - Single-Step Elevated Temperature Joining**
- **Summary/Conclusions**

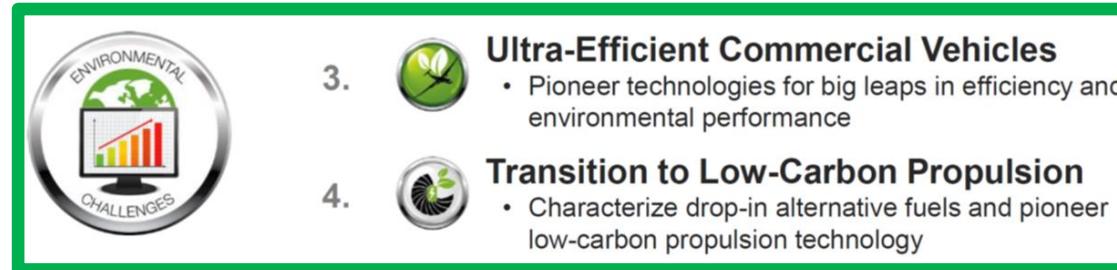
# Objectives

- ❖ **Deliver the benefits of ceramics and ceramic matrix composites (CMCs) for aerospace applications:**
  - Higher temperature capability.
  - Reduced cooling and weight.
  - Contributes to increased fuel efficiency, performance, range, and payload, and lower emissions and lower operation costs for future engines.
  
- ❖ **Develop joining and integration technologies**
  - Enable the wider utilization of ceramic matrix composite (CMC) turbine engine components by allowing for the fabrication of complex shaped CMC components and their incorporation within surrounding metal-based systems.



# CMC Turbine Engine Components and Joining Needs

## NASA Aeronautics Research Six Strategic Thrusts



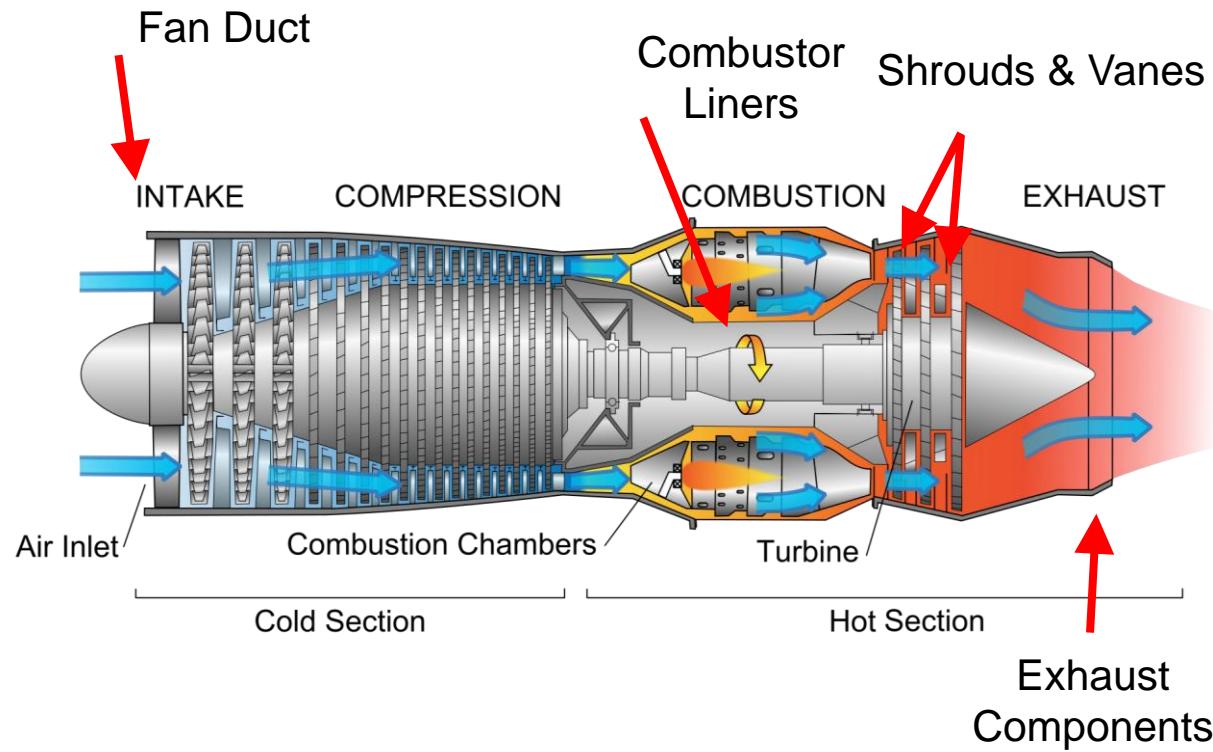
**Joining of singlet vanes to form doublets and joining of vane airfoils to ring sections (for smaller engines)**  
- Allows for a reduction in part count, seals, and leakage



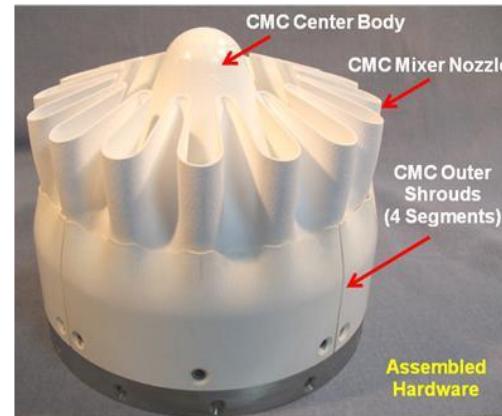
**Joining of airfoil and end caps**  
- Easier fabrication compared to a continuous 3-D CMC vane

# Components for Turbine Engine Applications

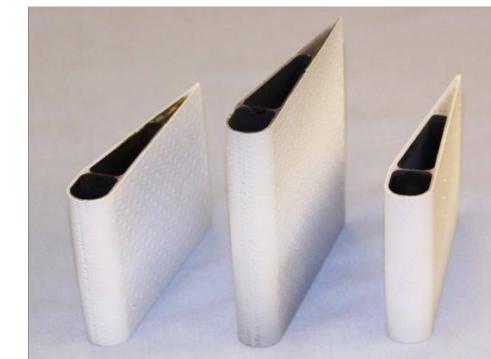
## Turbine Engines - Targeted Components (CMCs)



## NASA CMC Components



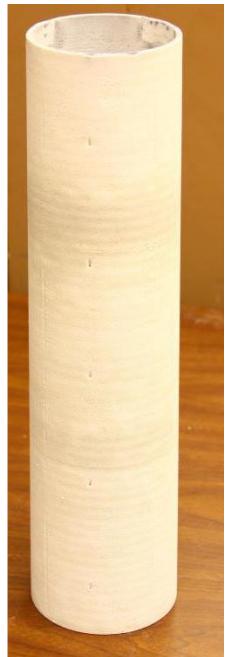
Oxide/Oxide Mixer Nozzle



EBC Coated SiC/SiC Vanes



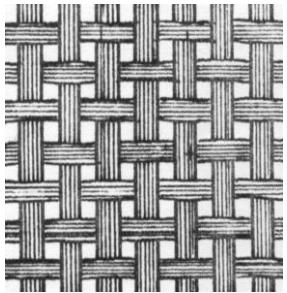
SiC/SiC Combustor Liners:  
Outer Liner and EBC  
Coated Inner Liner



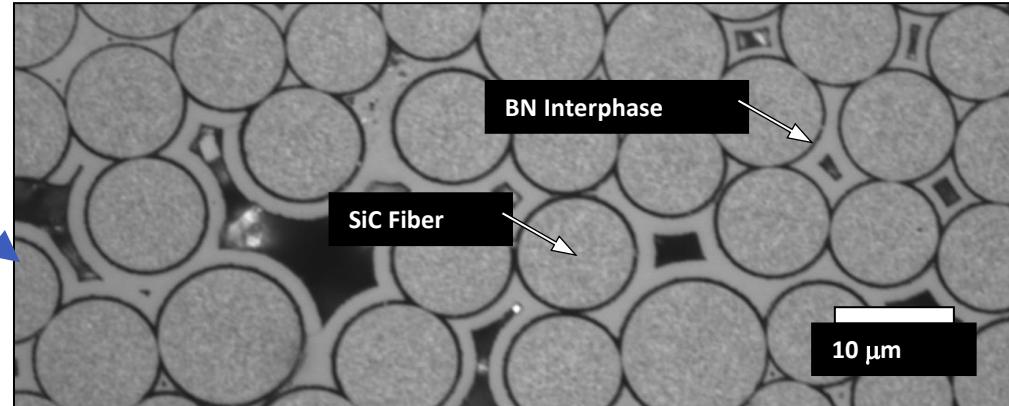
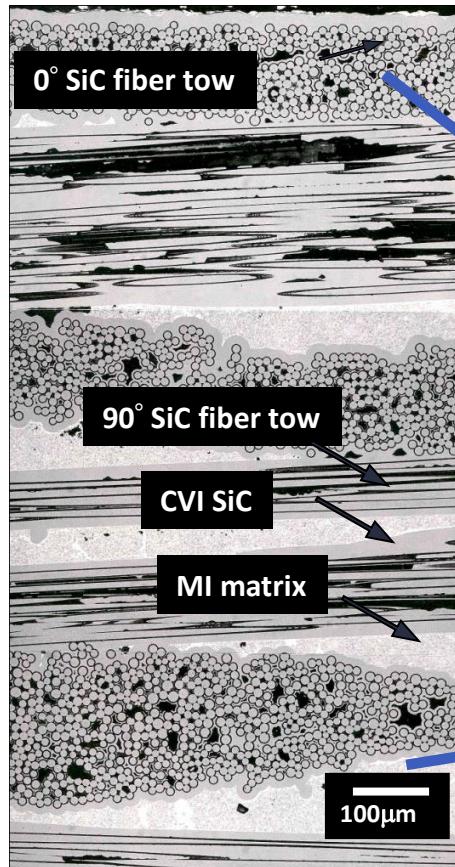
M.C. Halbig, M. Jaskowiak, J. Kiser, and D. Zhu. "Evaluation of ceramic matrix composite technology for aircraft turbine engine applications." In 51st AIAA Aerospace Sciences Meeting including the New Horizons Forum and Aerospace Exposition, p. 539. 2013.

# Conventional CMC Materials – Hybrid CVI and Melt Infiltrated (MI) SiC/SiC

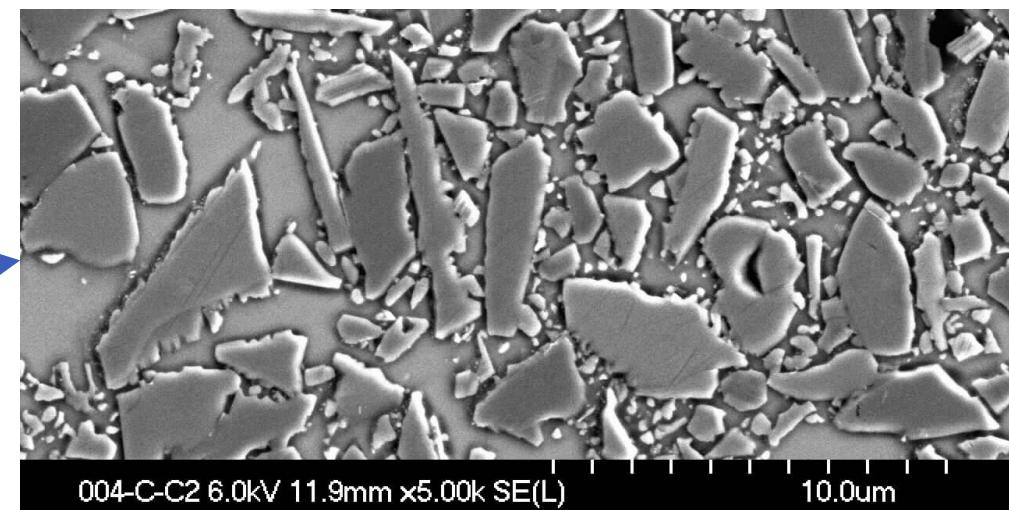
0-90 Plain Fiber Tow Weave



Composite Cross-Section



SiC fibers within a tow



SiC grains and silicon within MI matrix

- High thermal conductivity matrix
- Elimination of interlaminar porosity
- No matrix micro cracking
- Continuous fibers and fracture toughness



# Additively Manufactured SiC Fiber / SiC Matrix Composites Through Binder Jetting



ExOne Innovent



High pressure turbine cooled doublet vane sections.



Demo of a complex component, however the material is currently only suitable for lower stress applications.

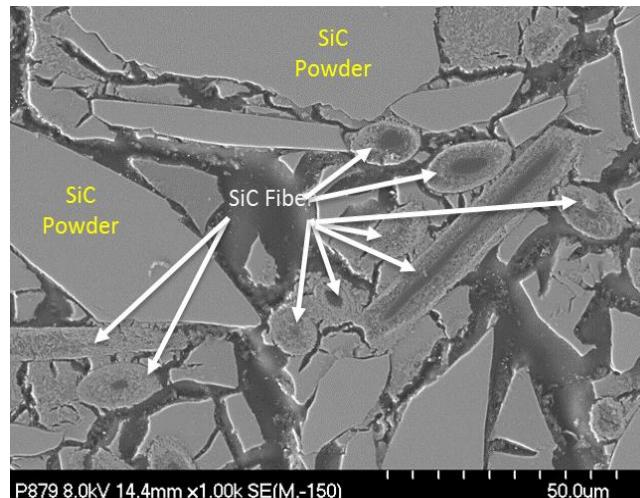
M.C. Halbig, J.E. Grady, M. Singh, J. Ramsey, C. Patterson, and T. Santelle. A Fully Nonmetallic Gas Turbine Engine Enabled by Additive Manufacturing of Ceramic Composites. No. E-19153. 2015.

## Constituents

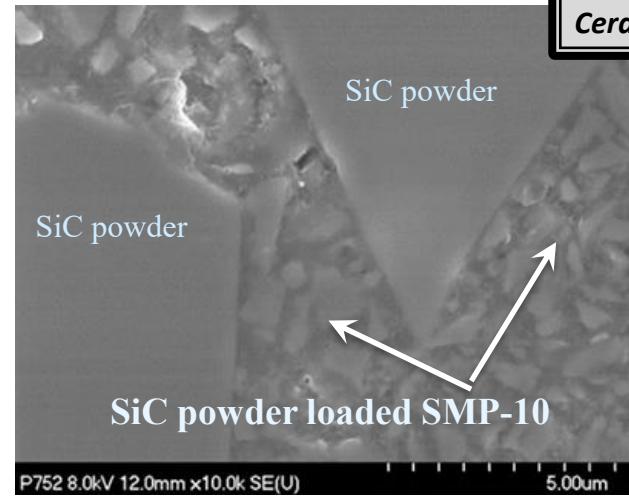


Si-TUFF iSiC fibers  
(Advanced Composite Materials, LLC)

~70  $\mu\text{m}$  long and  
~7  $\mu\text{m}$  in diameter

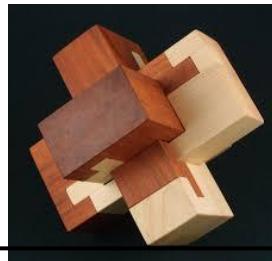


Short Fiber Reinforced Ceramic Matrix Composite

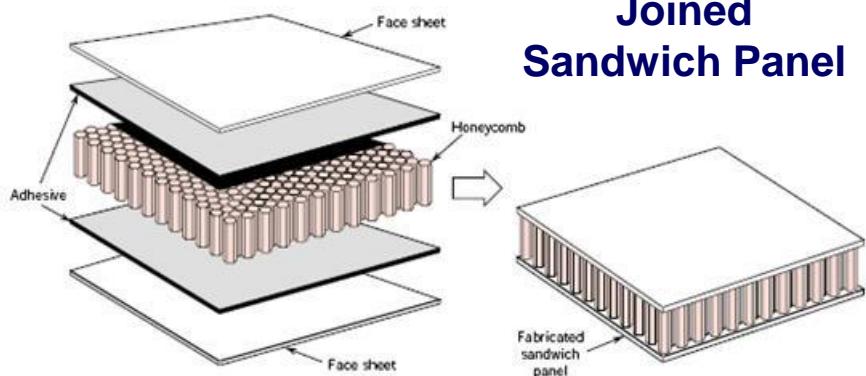


AM of fiber reinforced CMCs is in the early stages within the advanced processing field.

# Comparison of Layerwise Fabrication Methods for Ceramics

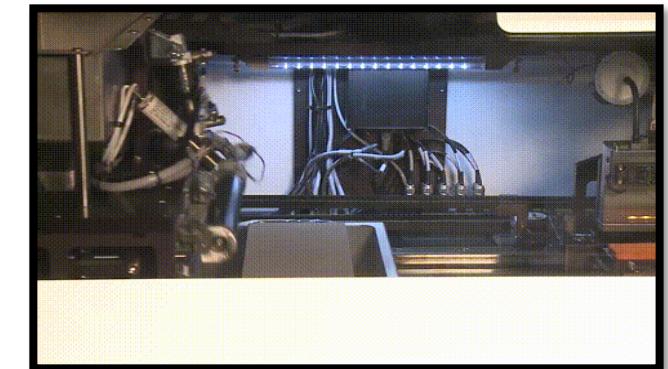
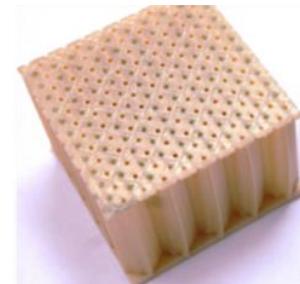


Characteristics	Joining	Additive Manufacturing
Waste compared to machining	Less	Less
Internal features	Yes	Yes
Part count	More	Less
Multiple fabrication steps	Yes	Depends
Ceramic Matrix Composite (CMCs)	Uses Conventional CMCs	Non-mature CMCs
Maturity Level	Relatively High	Relatively Low
Multimaterial structures	Easier	Harder
Structural ceramics	More Robust	Less Robust

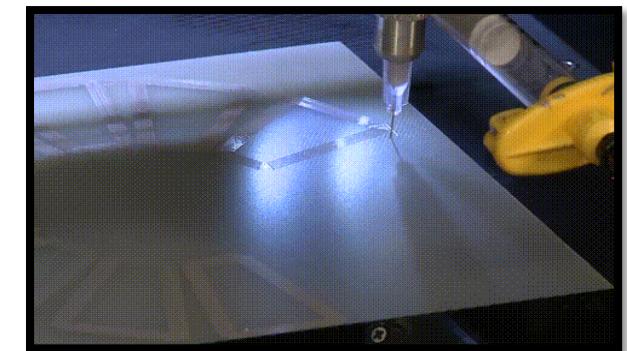


Joined  
Sandwich Panel

AM Polymer  
Sandwich Panel



Binder Jet 3D Printing



Direct Write Printing

AM is more challenging for fiber reinforced materials, multi-materials, co-processing, and robust structural ceramic parts.



# Joining and Integration of Ceramics and CMCs for Aerospace Applications

## Development Approach

- Develop single, multiple, and hybrid interlayer approaches to aid in the joining of ceramics and CMCs to themselves and to metals.
- Optimize processing conditions so that joints and parts remain strong and crack free.
- Investigate inter-relations between processing, microstructure, and properties.
- Evaluate the thermal and mechanical properties of the joint.
- Scale-up of processing to larger and more complex shaped sub-components.
- Evaluate joints in relevant conditions which are comparable to engine operating environments.

# Integration and Joining Technology Development

## **Ceramic to Ceramic (CMC to CMC) Systems:**

- **Brazing** - liquid metal flows into a narrow gap between the mating surfaces and solidifies to form a permanent bond. *Also for ceramic to metal joining.*
- **High Temperature Reactive Joining** - two step reactive formation of high temperature capable joints using carbon paste and Si infiltration (**ARCJoinT**).
- **Diffusion Bonding** - mating surfaces are pressed together and heated to cause bonding by interdiffusion of the components.
- **Refractory Eutectic Phase Bonding (REABond)** - melting of a eutectic phase from a solid to a single-phase liquid.
- **Single-step Elevated Temperature Joining (SET)** - single step reaction formed SiC joint for  $>2400^{\circ}\text{F}$  applications using C, Si, and SiC-based slurries.

**Uniform, dense, crack-free joints from all approaches.**



# Comparison of CMC Joining Approaches

Characteristics	Joining Approach				
	Brazing (Cu-Si-Ti based)	ARCJoinT	Diffusion Bonding (Ti)	REABond (Si-Hf)	SET Joining (C,Si,SiC based)
Temperature limit	<1472°F (800°C)	<2400°F (1316°C)	~2373°F (1300°C)	<2400°F (1316°C)	>2400°F (1316°C)
Little or no processing pressure	√	√	✗	√	√
Curved shape joining	√	√	✗	√	√
Simple, one-step processing	√	✗	√	√	√
Substrate surface condition	smooth or rough	smooth or rough	smooth	smooth or rough	smooth or rough
Ceramic or metal joining	both	ceramic	ceramic	ceramic	ceramic
Interlayer type	foils, pastes	pastes	foils, surface coatings	pastes, tapes	pastes, tapes
Cure CMC processing flaws (e.g. porosity and microcracks)	✗	√	✗	√	√
Issues	possible formation of brittle ceramic phases	free silicon	geometry limitations and processing stress	silicon rich phase	early in development
Bond quality	uniform, dense, and crack-free joints	uniform, dense, and crack-free joints	uniform, dense, and crack-free joints	uniform, dense, and crack-free joints	uniform, dense, and crack-free joints

Several joining approaches are available for various material pairings and application requirements.

# Diffusion Bonding, REABond, and Brazing Joining Processes



## Materials (dimensions 0.5" x 1")

- Substrates: CVD SiC, SA-Tyrannohex (parallel), and SA-Tyrannohex (perpendicular).
- **Interlayers:** Ti foil (10, 20 micron) and B-Mo alloy foil (25 micron)

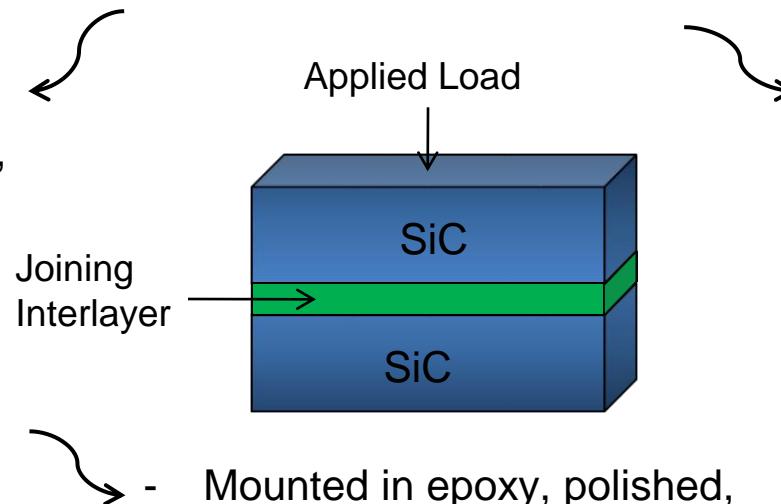
## Diffusion Bonding

- Atmosphere: Vacuum
- Temperature: Ti 1200°C, B-Mo 1400°C
- **Pressure: 30MPa**
- **Duration: 1-4 hr**
- Cool down: 2 °C/min

## Material definitions:

CVD SiC => chemically vapor deposited SiC

SA-Tyrannohex => Woven SA-Tyranno fiber hot pressed composite like material



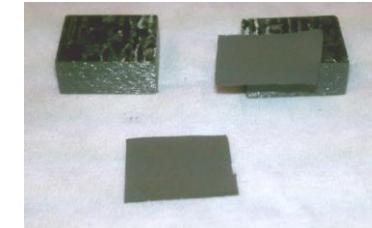
- Mounted in epoxy, polished, and joints characterized using optical microscopy and scanning electron microscopy with energy dispersion spectroscopy analysis.
- Thermomechanical analysis

## Materials (dimensions 0.5" x 0.5")

- CMC materials: C/C, MI SiC/SiC, CVI SiC/SiC, prepreg MI SiC/SiC, and SA-Tyrannohex.
- **Interlayer:** Si-Hf Eutectic tapes of 1, 2, and 3 layers.
- **Brazes:** single layer, multi-layer, and tailored.

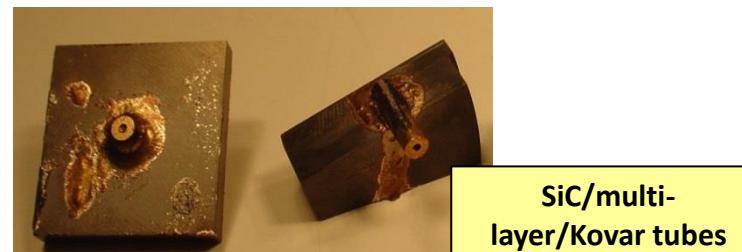
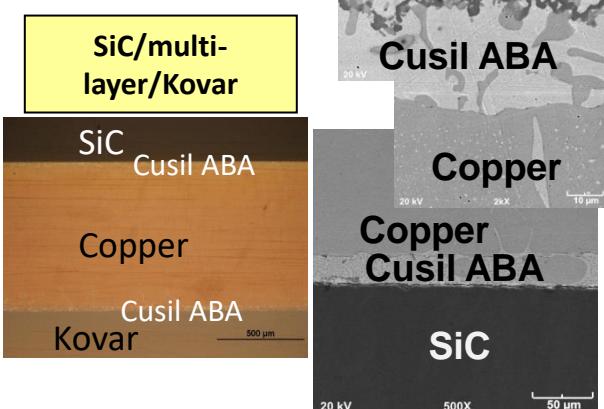
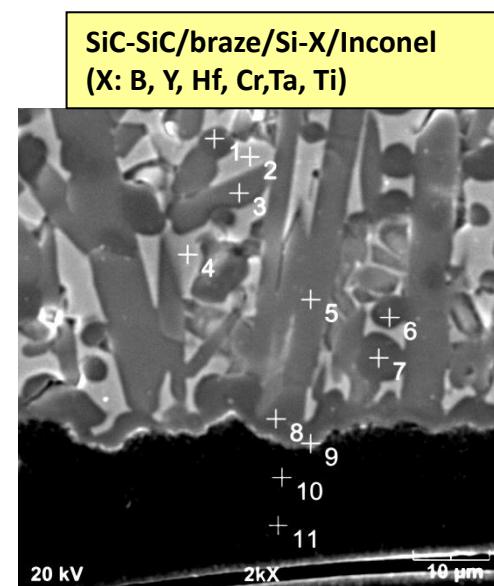
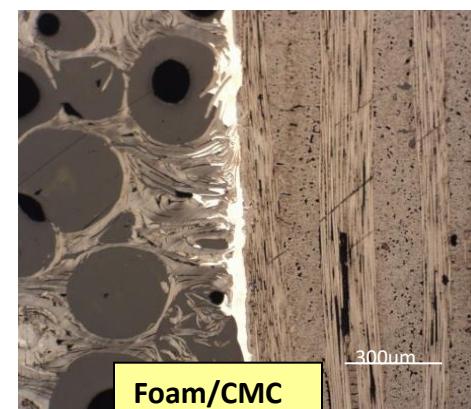
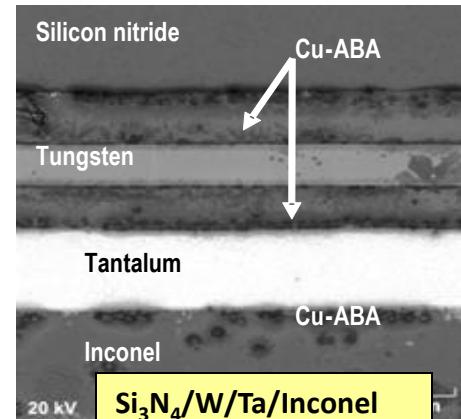
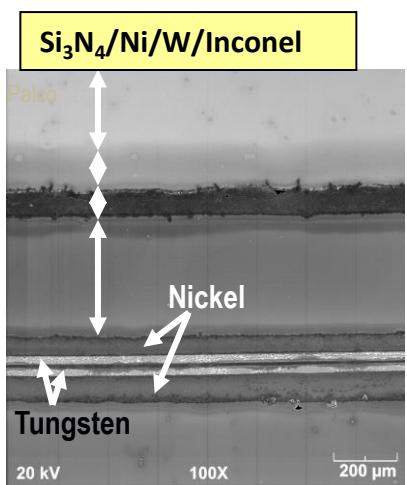
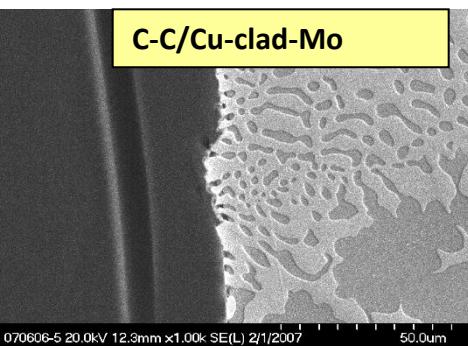
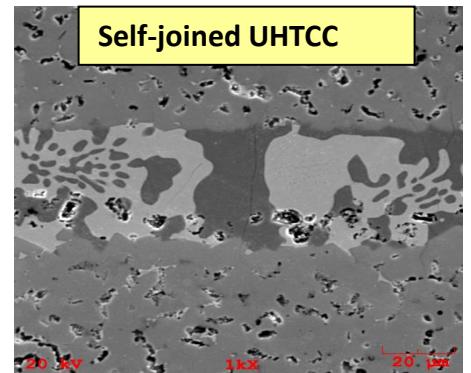
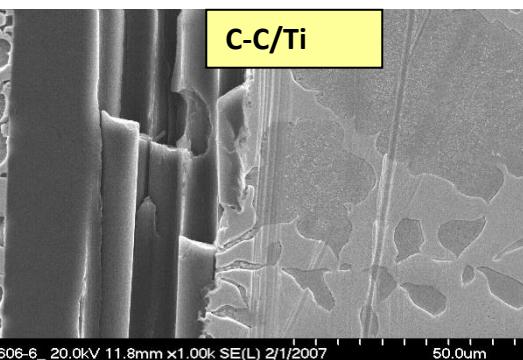
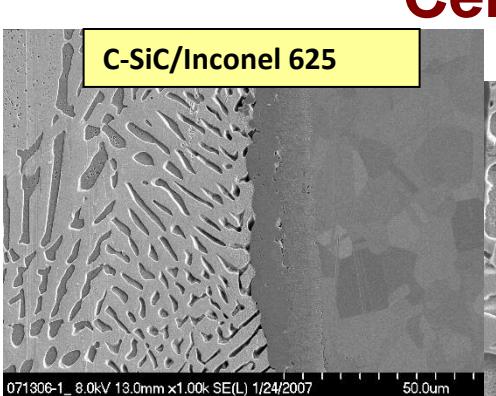
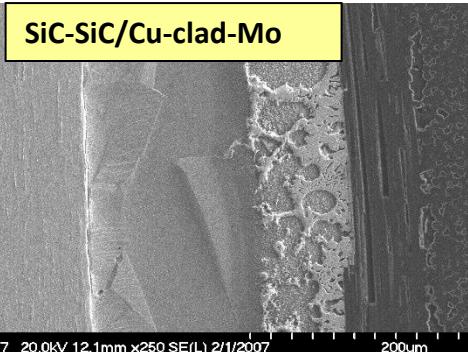
## REABond and Brazing

- Atmosphere: Vacuum
- Temperature: Reabond: 1340°C  
Brazing ~10°C above the braze liquidus temperature.
- **Load: 100 g/sample**
- **Duration: 10 minutes**
- Cool down: 2 °C/min



Joining prep with CMC substrates and Si-Hf REABond tapes with 30-35% solid loading.

# Brazing: Ceramic to Ceramic and Ceramic to Metal Systems

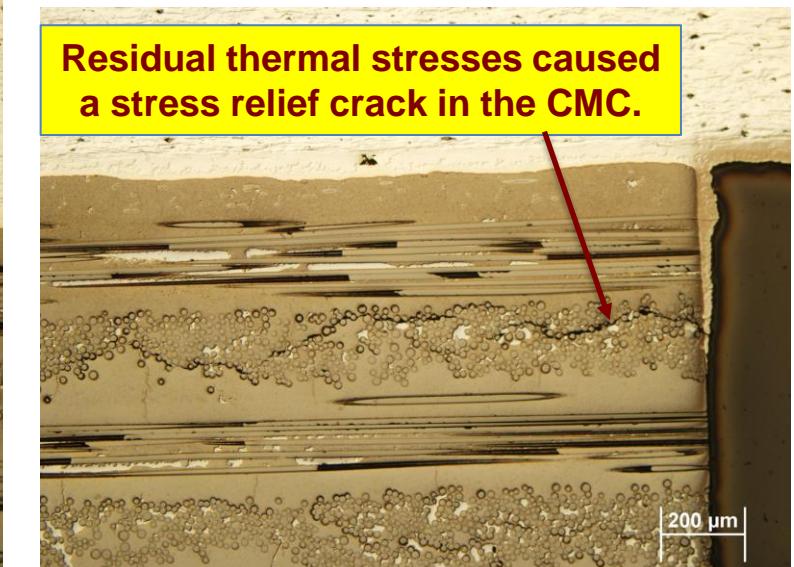
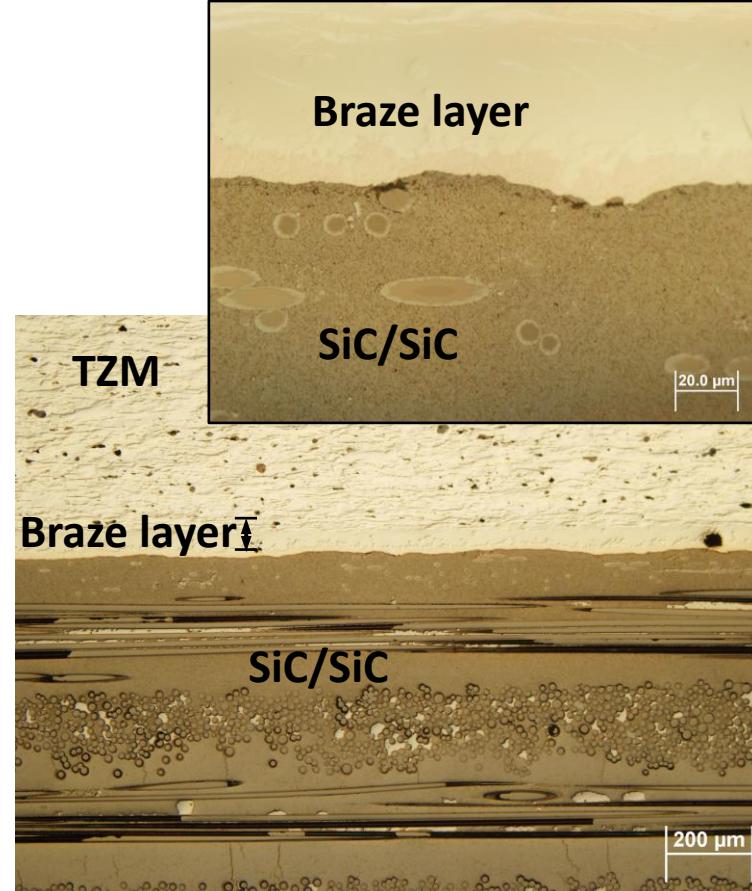
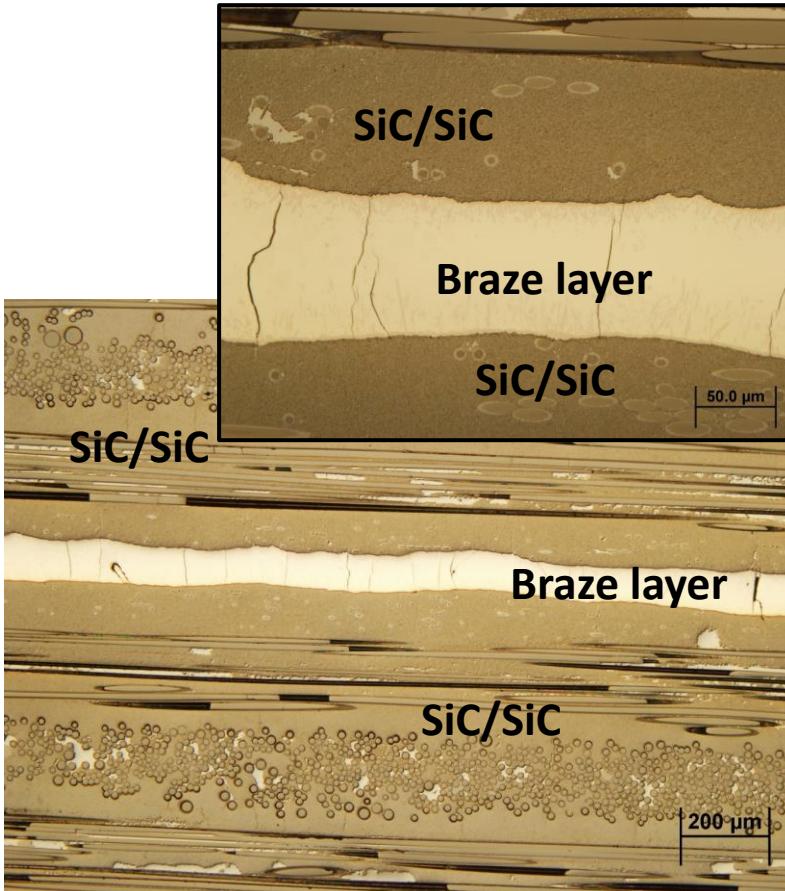


**Design and Understanding of Interfaces is Key to Successful Integration**

**Successful brazing with many material combinations.**

# Brazing: CMCs to CMCs and to Metals

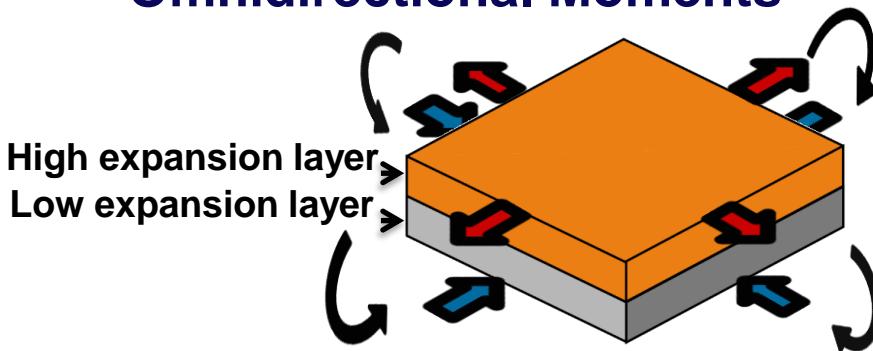
- Brazing of SiC/SiC to SiC/SiC and TZM
- TiBraze 200 foil: Ti(40)-Zr(20)-Cu(20)-Ni(20) in wt. %



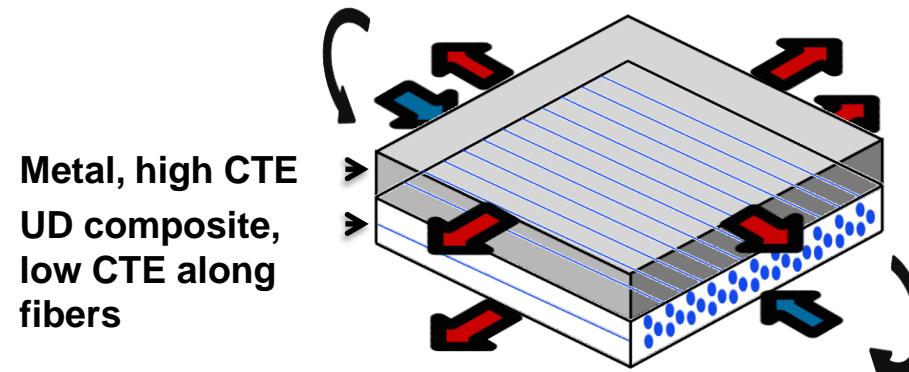
Good wetting was seen on both substrates: SiC/SiC and TZM.

# Brazing: Integration of Metals to CMCs for Thermally-Actuated, High Temp. Morphing Composites

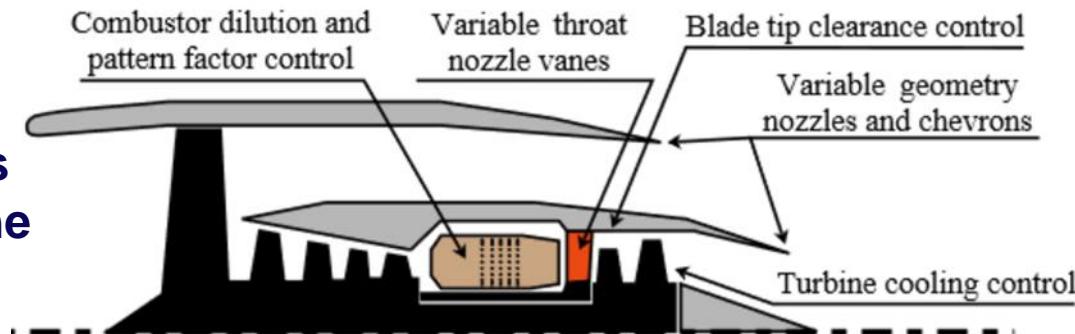
## Isotropic Bimorph- Omnidirectional Moments



## Composite Construction Allows General Platforms

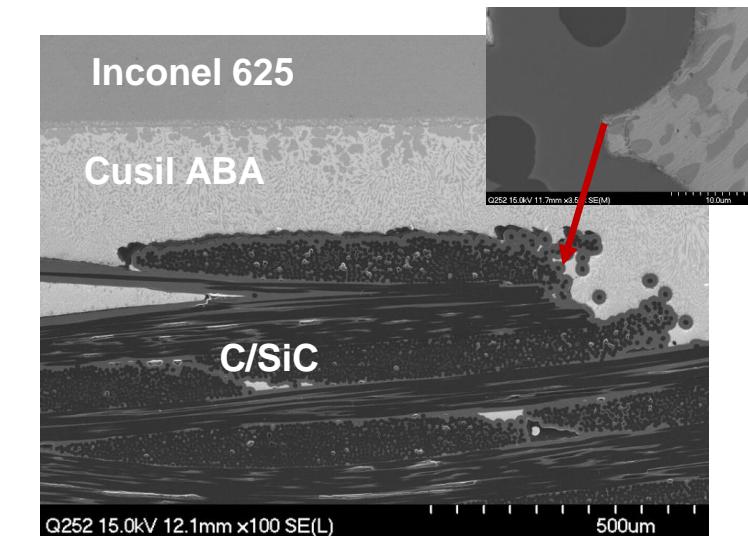


## Potential Gas Turbine Engine Applications



Morphing of CMC-metallic flap at 1000°C (yellow) compared to R.T.

E. Eckstein, M.C. Halbig, and P. Weaver,  
"Thermally-driven morphing with high  
temperature composites." In *57th  
AIAA/ASCE/AHS/ASC Structures,  
Structural Dynamics, and Materials  
Conference*, p. 1241. 2016.



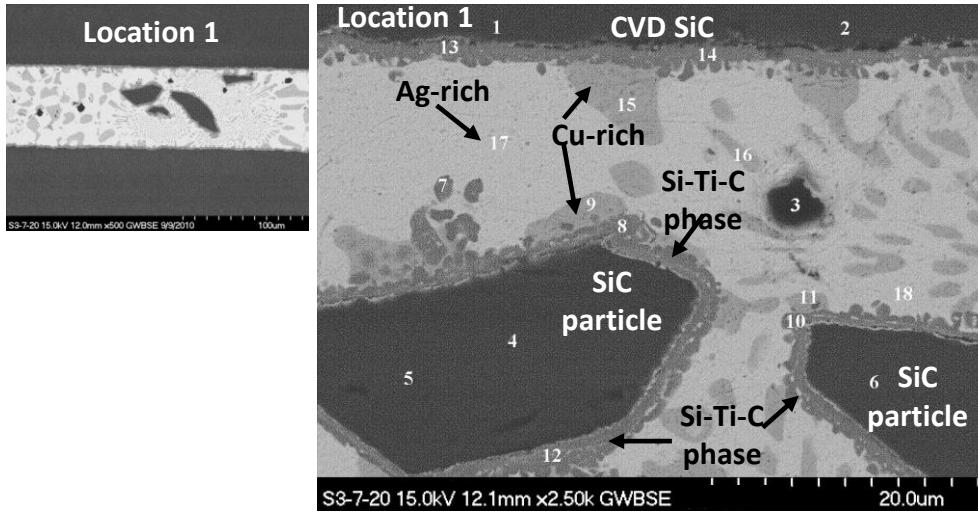
## Brazing of C/SiC to Inconel

Bolted flap demonstrated and brazing initiated for fully integrated concepts.

# Brazing: Interlayer Property Modifications - SiC Particulate Additions to Ticusil Brazing Paste for CVD SiC to CVD SiC Joining

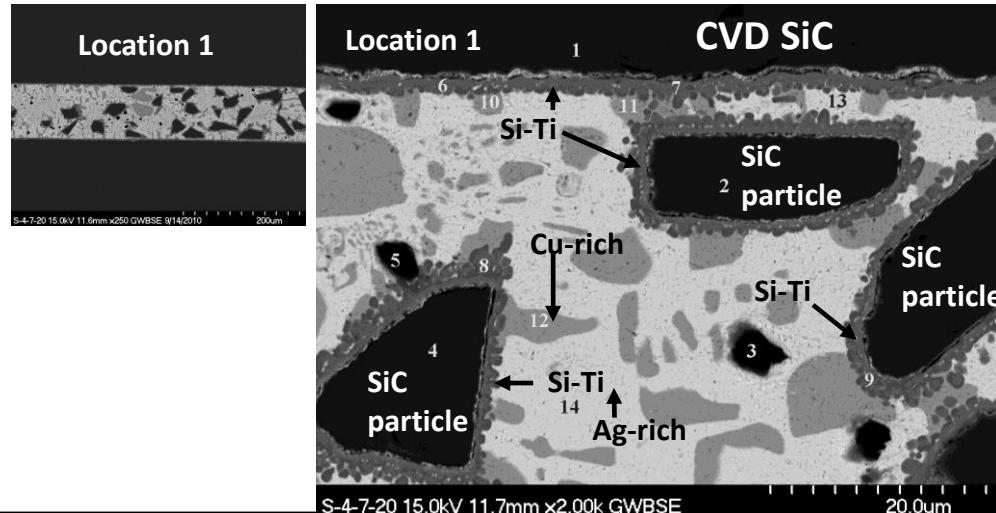


## CVD SiC/Ticusil (10wt% SiCp)/CVD SiC



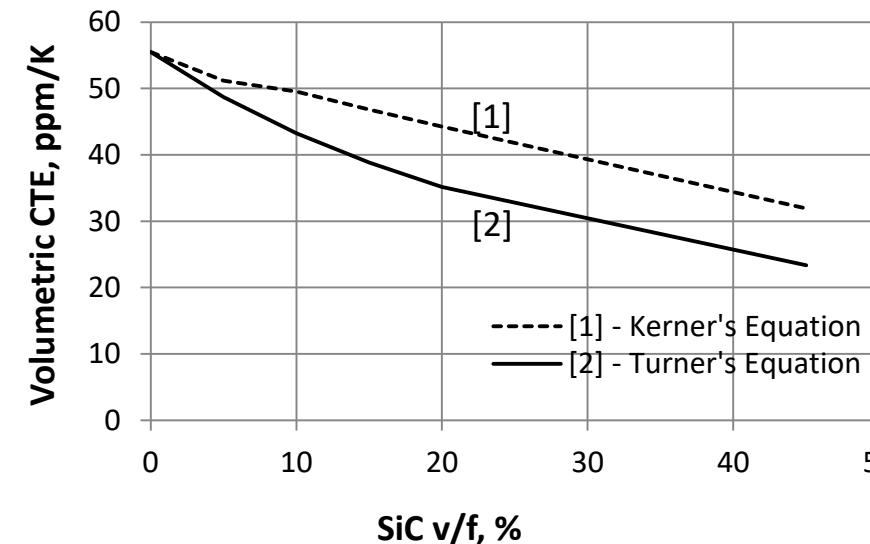
**Ticusil composition in wt. %:**  
68.8% Ag, 26.7% Cu, and 4.5% Ti

## CVD SiC/Ticusil (15wt% SiCp)/CVD SiC



	Ticusil Paste			
	0 wt% SiCp	5 wt% SiCp	10 wt% SiCp	15 wt% SiCp
	$\mu \pm \sigma$	$\mu \pm \sigma$	$\mu \pm \sigma$	$\mu \pm \sigma$
CVD SiC	3442 $\pm$ 71	3304 $\pm$ 86	3134 $\pm$ 117	3305 $\pm$ 119
Braze	252 $\pm$ 58	86 $\pm$ 5	117 $\pm$ 52	106 $\pm$ 31
CVD SiC	3286 $\pm$ 71	3287 $\pm$ 95	3241 $\pm$ 51	3239 $\pm$ 111

Mean ( $\mu$ ) & Standard Deviation ( $\sigma$ ) HK of Ticusil Joints



Predicted effect of SiC reinforcement on the volumetric CTE of Ticusil (or Cusil-ABA) braze.

**Affects of particulate additions:**

- Pulled Ti out of the braze
- Formed Ti-Si-C phases at SiC surfaces
- Decreased the hardness of the braze
- Predicted to lower the volumetric CTE by 40-60% with 40 vol% SiCp.

# ARCJoinT: Joining of Ceramic Components Using Affordable, Robust Ceramic Joining Technology (ARCJoinT)

Apply Carbonaceous Mixture to Joint Areas

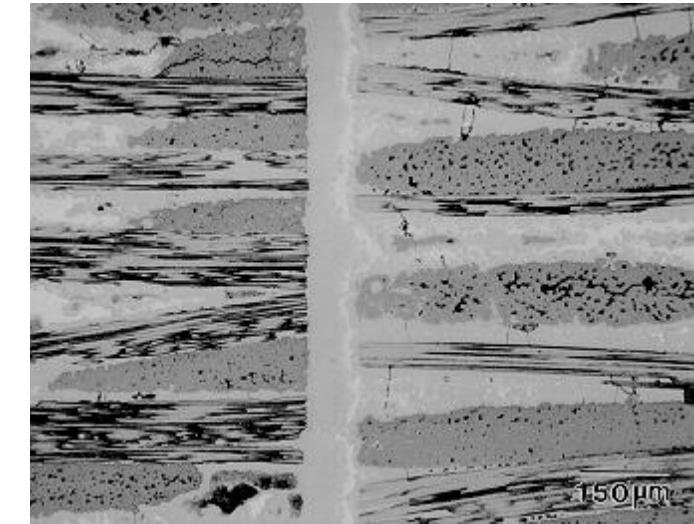
Cure at 110-120°C for 10 to 20 minutes

Apply Silicon or Silicon-Alloy (paste, tape, or slurry)

Heat at 1250-1425°C for 10 to 15 minutes

Affordable and Robust Ceramic Joints with Tailorable Properties

1999 R&D 100 Award  
2000 NorTech Innovation Award (M. Singh)



Joined MI C/SiC Composite

## Advantages

- Joint interlayer properties are compatible with parent materials.
- Processing temperature around 1200-1450°C.
- No external pressure or high temperature tooling is required.
- Localized heating sources can be utilized.
- Adaptable to in-field installation, service, and repair.

Very good quality, high strength bonds are obtained. However, the joining method requires a two-step process and is limited to temperatures <2400°F (1316°C).

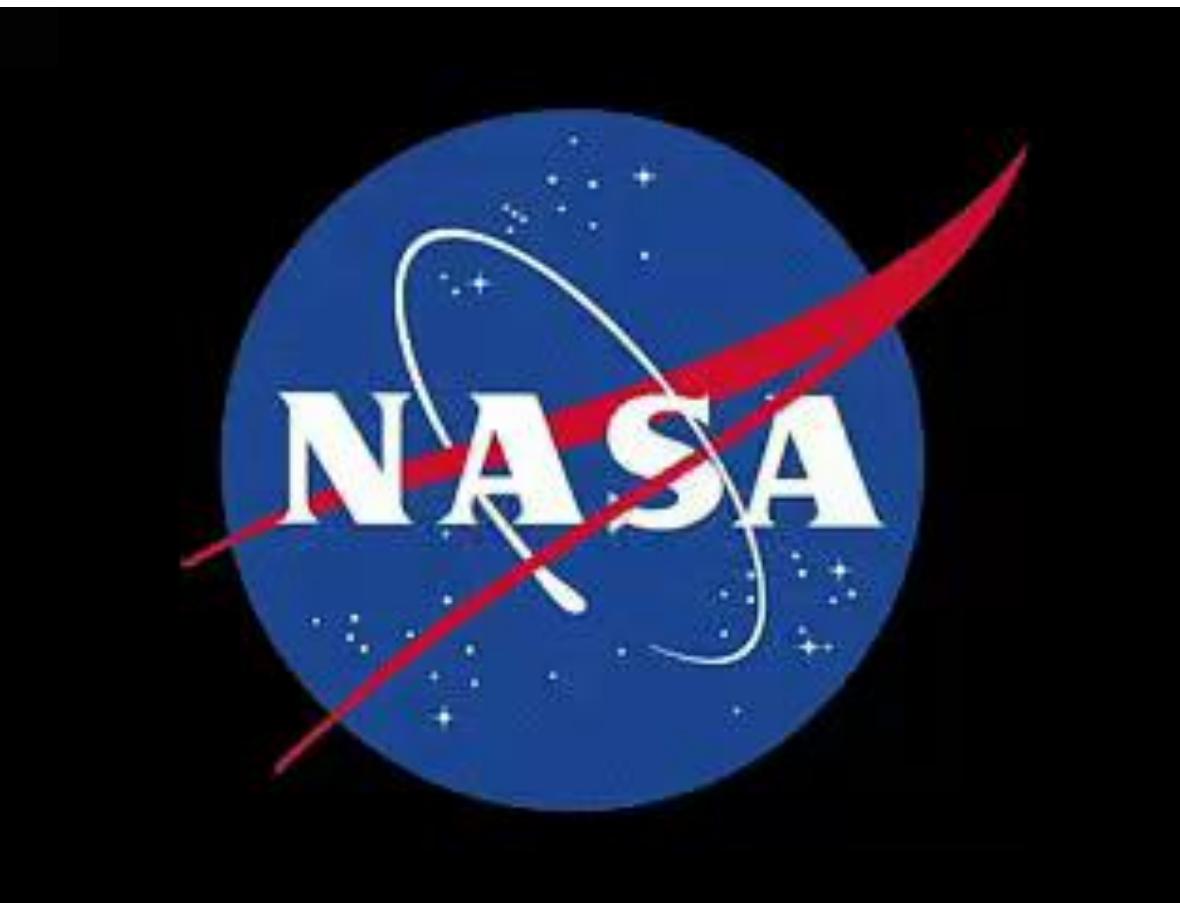


# Diffusion Bonding and Brazing: Fabrication of Lean Direct Injector Components

Initial effort by a different group was unsuccessful:

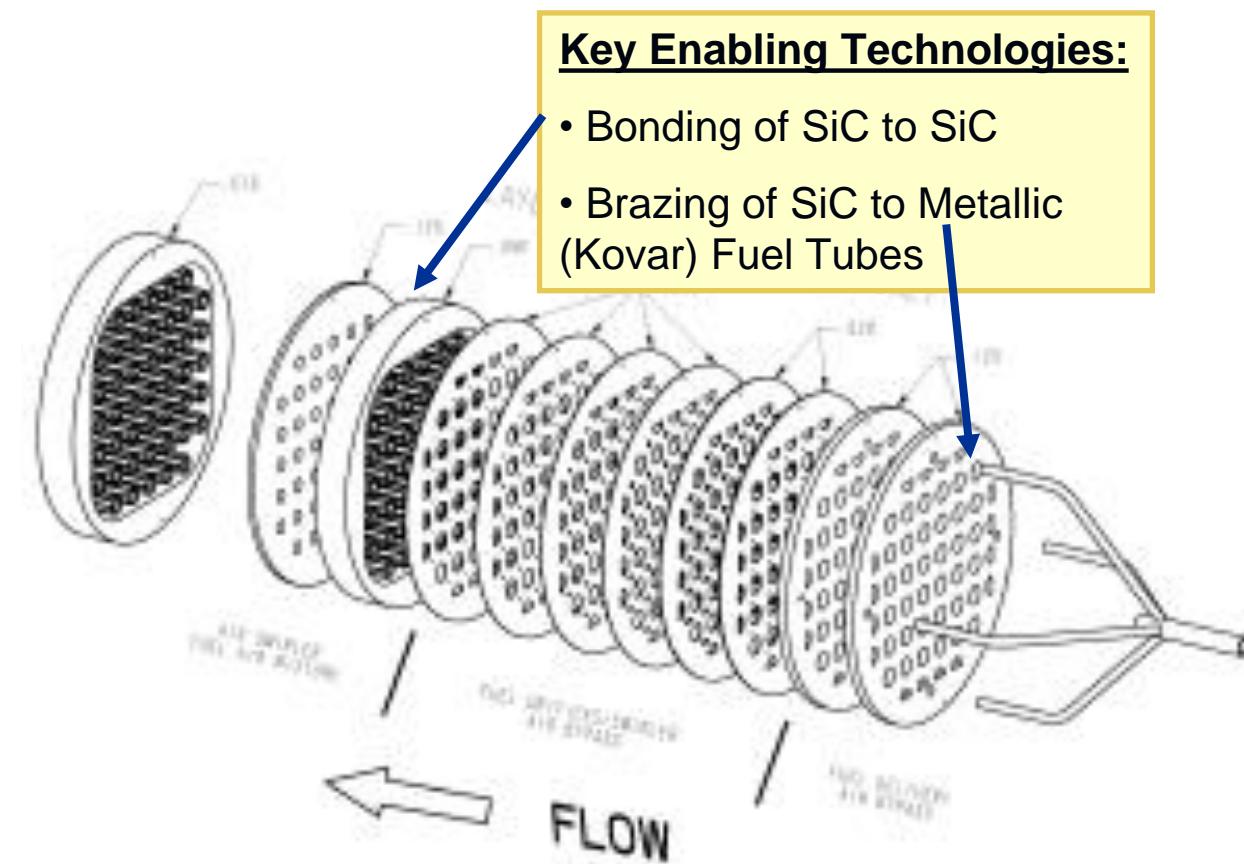
## Disadvantages of Joining Silicon Carbide with a Silicate Glass Layer

- Difficult to achieve a uniform layer
- Relatively low strength
- Glass flows and fills in holes and edges where it is not desired
- Glass joints were not leak-free



## Benefits of Laminated Plates

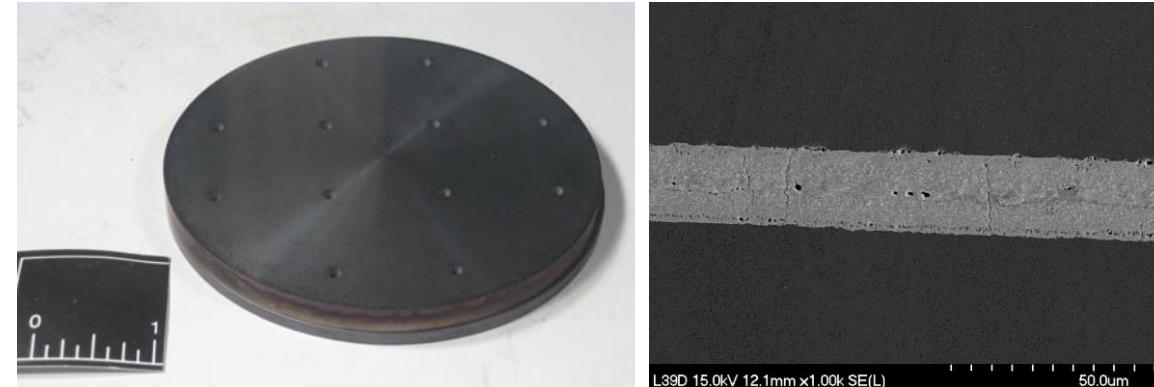
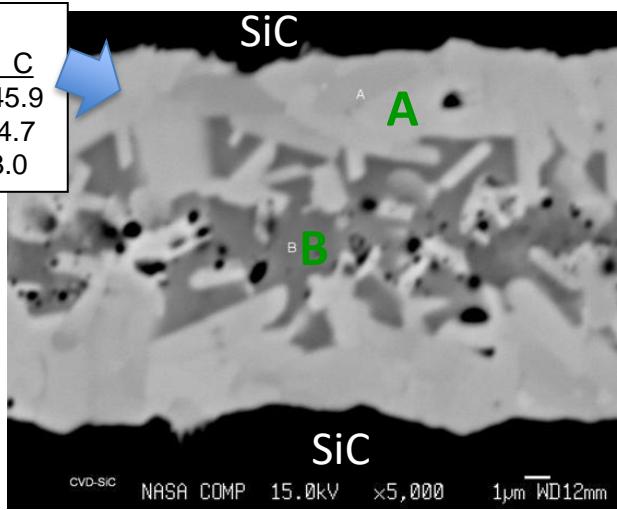
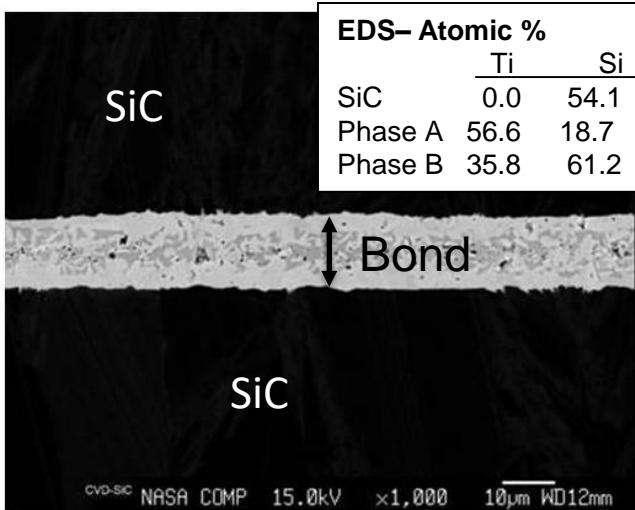
- Passages of any shape can be created to allow for multiple fuel circuits
- Provides thermal protection of the fuel to prevent choking
- Low cost fabrication of modules with complicated internal geometries through chemical etching



# Diffusion Bonding: CVD SiC to CVD SiC with PVD Ti for Lean-Direct Injector Fabrication

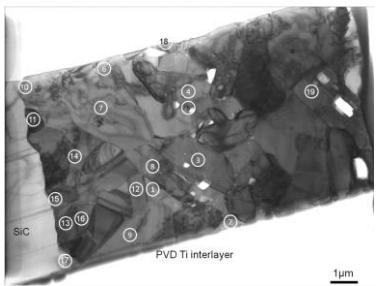
10  $\mu\text{m}$  Interlayer (1200°C, 30MPa, 2 hr, vacuum, cool down 2 °C/min)

## SEM and EDS

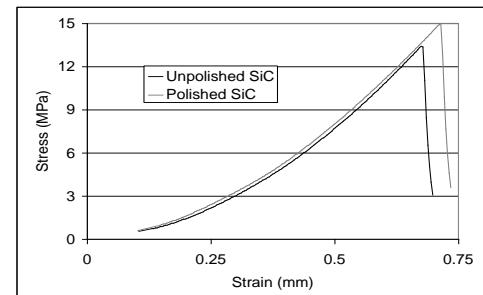
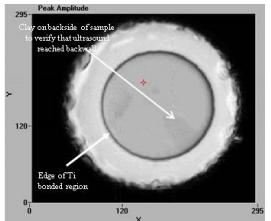


Optimized diffusion bonding conditions were applied to injector sub-elements.

## TEM w/SAD



## Ultrasonic Immersion and Pull Tests



Strengths of 15.0 MPa were twice the requirement.

- Very good quality bonds are obtained that are uniform and crack free.
- The joining process requires high applied loads and flat sub-elements for joining.
- However, diffusion bonding is well suited for injector fabrication.
- The PVD Ti coating w/diffusion bonding provides a non- flowing joining approach.

M.C. Halbig, M. Singh, and H. Tsuda. "Integration technologies for silicon carbide-based ceramics for micro-electro-mechanical systems-lean direct injector fuel injector applications." *International Journal of Applied Ceramic Technology* 9, no. 4 (2012): 677-687.

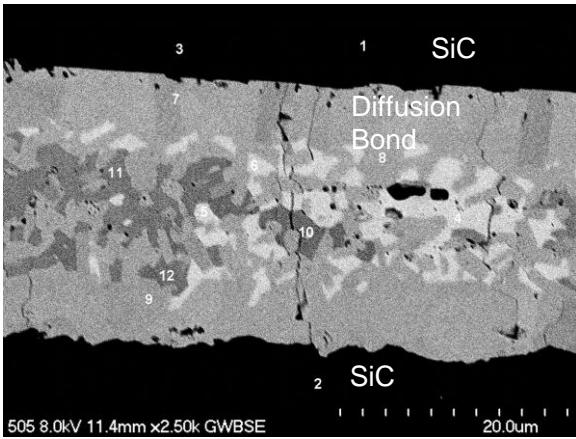


Topical issue on  
"Ceramic Integration  
Technologies,"  
July/August 2012 Issue

# Diffusion Bonding: CVD $\beta$ -SiC Substrates with Interlayers of Metallic Titanium Foils and PVD Titanium

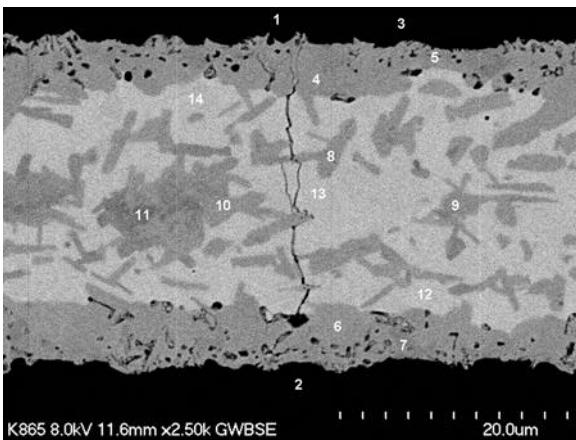
## PVD Ti Interlayer at 1250°C

20 micron interlayer and a 2 hr hold

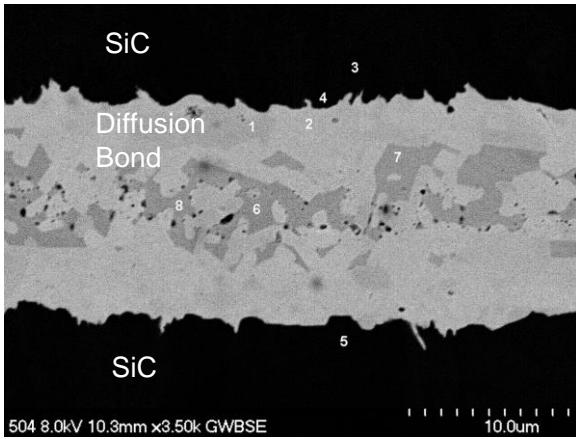


## Ti Foil Interlayer at 1200°C

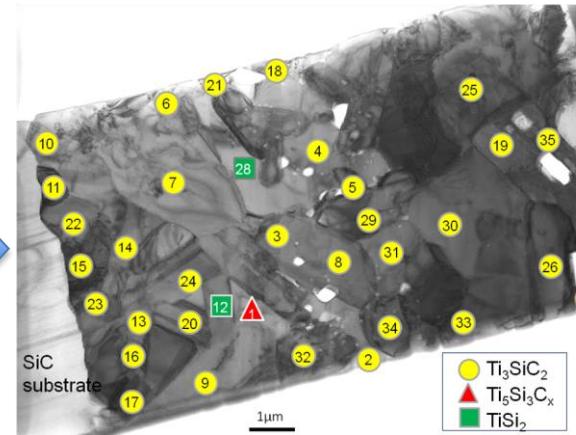
20 micron foil and a 2 hr hold



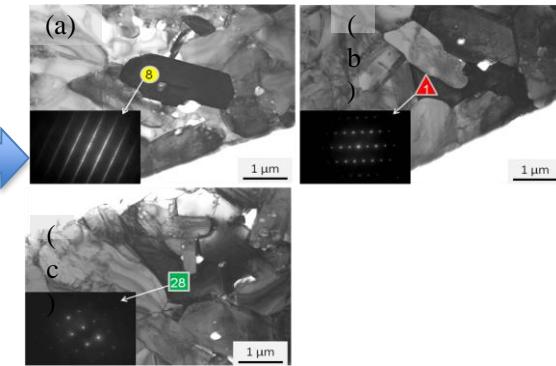
10 micron interlayer and a 2 hr hold



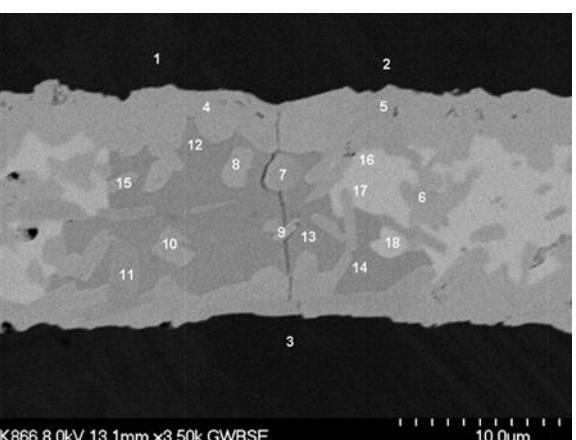
TEM micrograph and determined phases



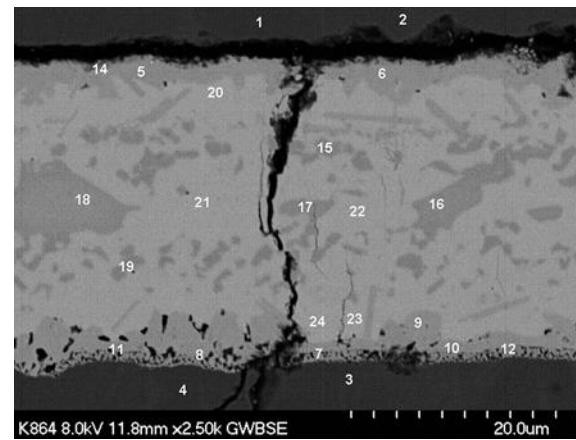
TEM micrographs and SAD patterns:  
(a)  $\text{Ti}_3\text{SiC}_2$  ( $B = [1120]$ ), (b)  $\text{Ti}_5\text{Si}_3\text{C}_x$  ( $B = [7253]$ ), (c)  $\text{TiSi}_2$  ( $B = [111]$ ).



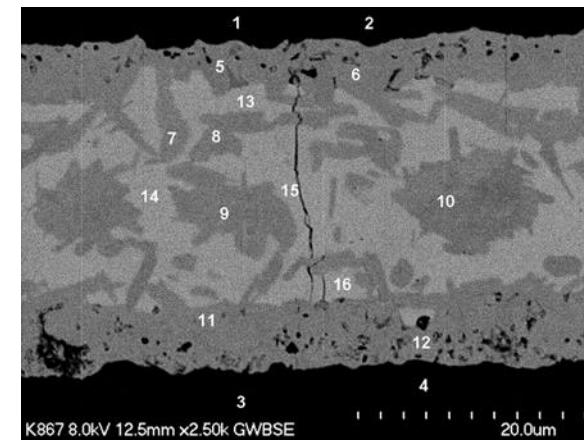
10 micron foil and a 2 hr hold



20 micron foil and a 1 hr hold



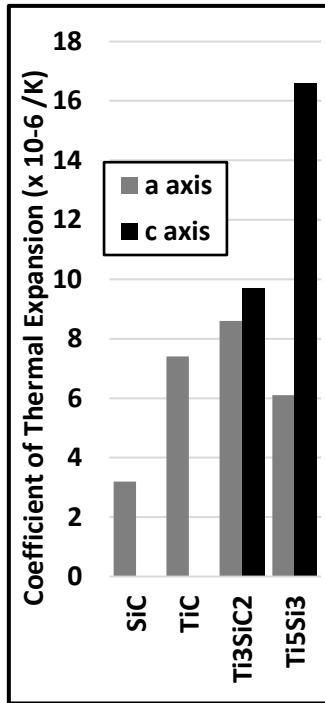
20 micron foil and a 4 hr hold



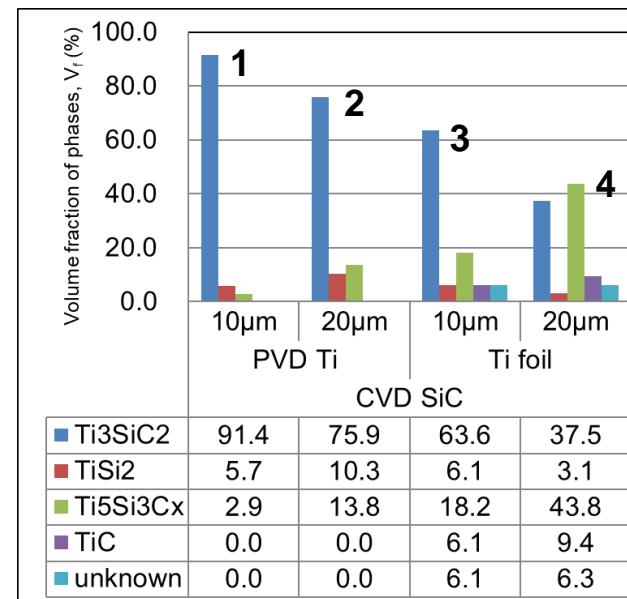
M.C. Halbig, M. Singh, H. Tsuda, and R. Asthana. "Diffusion bonding of SiC ceramics with interlayers of metallic titanium foils and PVD titanium coatings." *International Journal of Applied Ceramic Technology* (2022).

Thinner Ti layers and/or longer processing time can react away intermediate phases and provide crack free or minimally cracked bonds.

# Diffusion Bonding: CVD $\beta$ -SiC Substrates with Interlayers of Metallic Titanium Foils and PVD Titanium

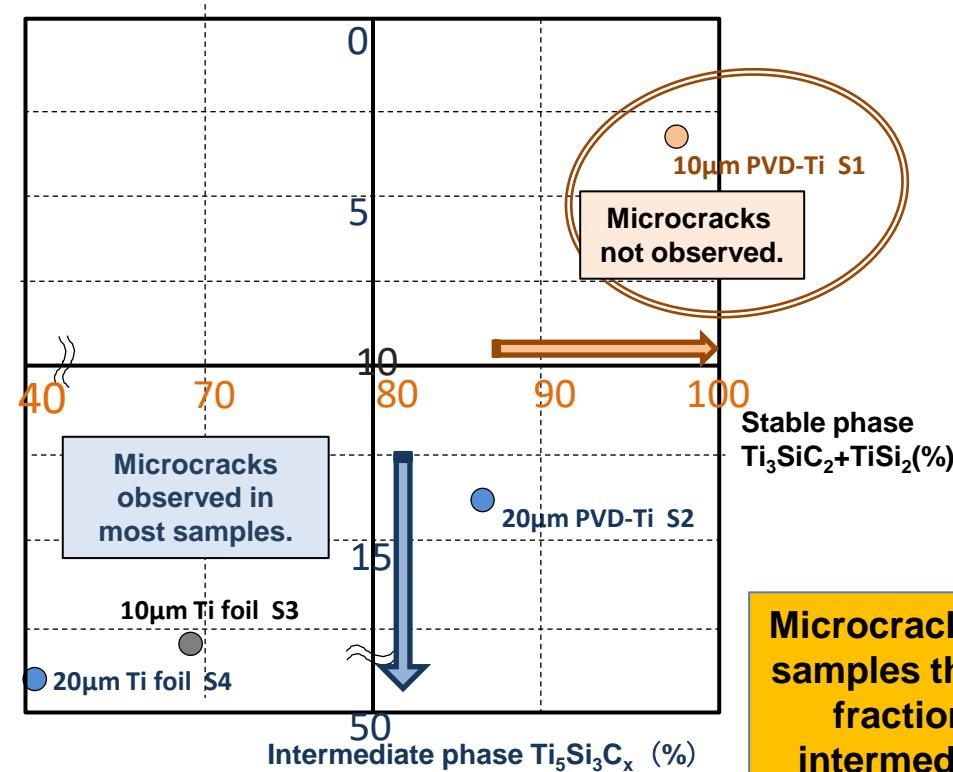


CTEs of the phases.



From TEM analysis, calculated fractions of phases (all 2 hours).

$\text{Ti}_5\text{Si}_3\text{C}_x$  ( $\text{Ti}_5\text{Si}_3$ ) is highly anisotropic in its thermal expansion where  $\text{CTE}(\text{c})/\text{CTE}(\text{a}) = 2.72$  (Schneibel et al). Naka et al suggest this is an intermediate phase.

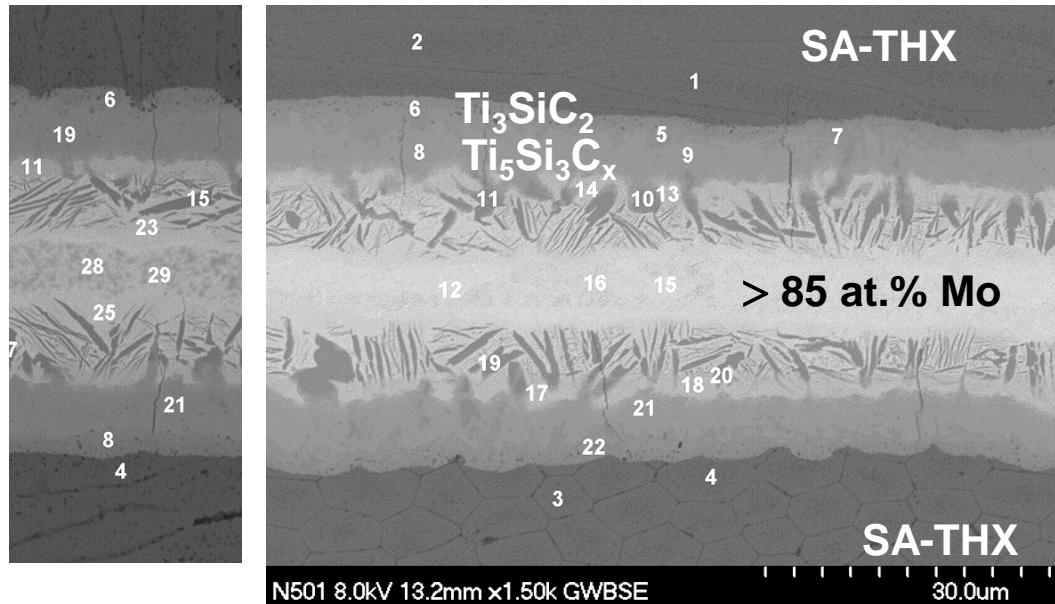


Observed relationship between phase grain fraction and crack formation

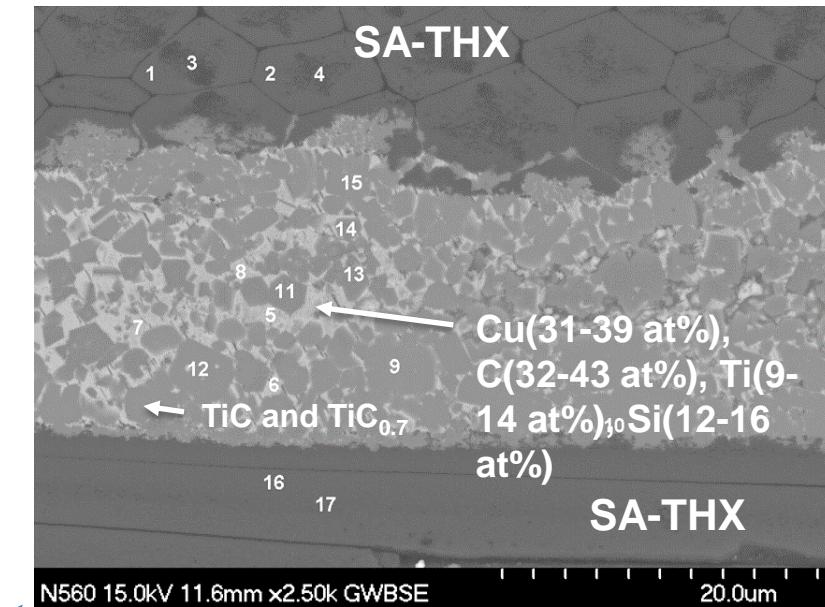
Very good quality bonds are obtained that are uniform and crack free. However, the joining process requires high applied loads and flat sub-elements for joining.

# Diffusion Bonding of SiC Fiber-Bonded Ceramics, SA-Tyrannohex™, using Ti/Mo/Ti and Ti/Cu/Ti Interlayers

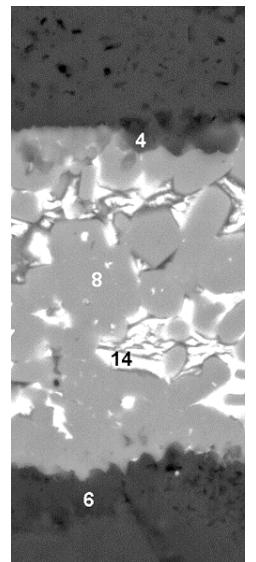
⊥SA-THX    SA-THX w/ fibers || to joining plane



SA-THX w/ fibers || to joining plane



⊥SA-THX

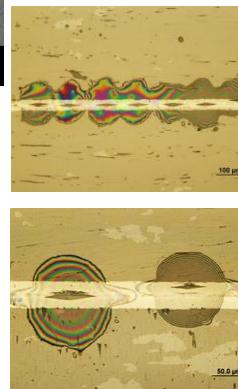


## Ti/Mo/Ti Diffusion Bond Layers:

- Lowered Mo bonding temp. from 1500°C to 1200°C (30MPa, 3 hr)
- Introduced a ductile interlayer (half the hardness at the center versus the edge).



## Microhardness testing



## Ti/Cu/Ti Diffusion Bond Layers:

- Cu layer seems to accelerate diffusion and reaction kinetics.
- No micro-cracking as for only a Ti layer where  $Ti_5Si_3Cx$  has formed. However, shrinkage cavities are observed.

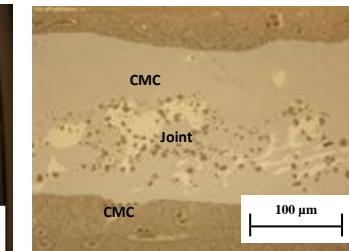
M.C. Halbig, R. Asthana, and M. Singh. "Diffusion bonding of SiC fiber-bonded ceramics using Ti/Mo and Ti/Cu interlayers." *Ceramics International* 41, no. 2 (2015): 2140-2149.

Multi-layers can be used to decrease bonding temperature, increase reaction kinetics, tailor properties, and reduce micro-cracking.

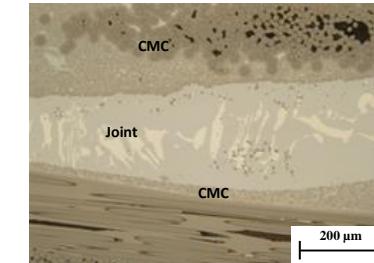
# REABond: Joining Various CMCs with Two Si-8.5Hf Eutectic Tapes [210 microns each]



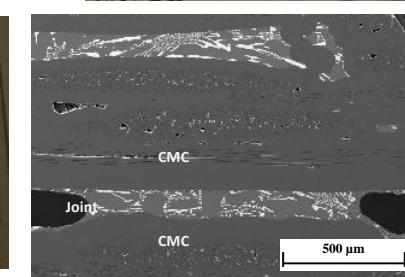
GE  
Hypercomp II  
MI SiC/SiC



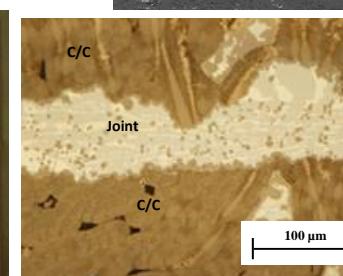
BFG MI  
SiC/SiC



Hyper-  
Therm CVI  
SiC-SiC



Goodrich  
3-D C/C  
(3 tapes)

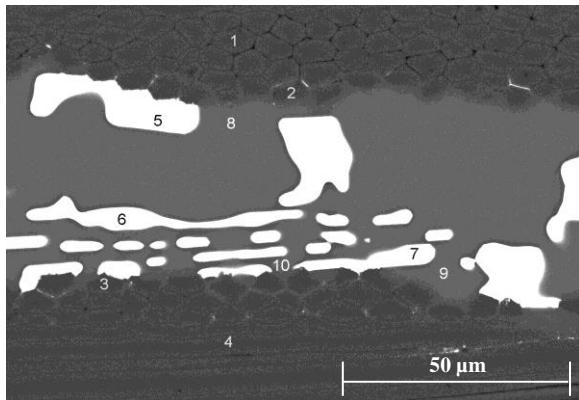


Porous CMCs provide extra challenges by depleting the interlayer material at joining gaps (matrix is infiltrated).

# REABond: Joining of SA-Tyrannohex and Mechanical Testing by Single Lap Offset (SLO)



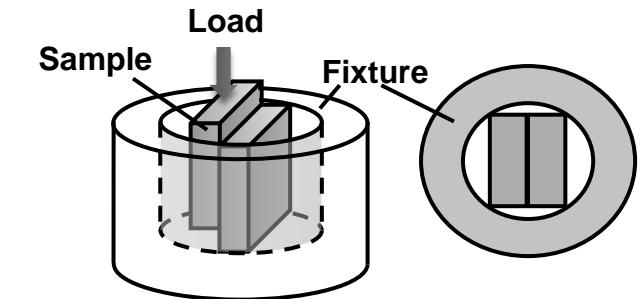
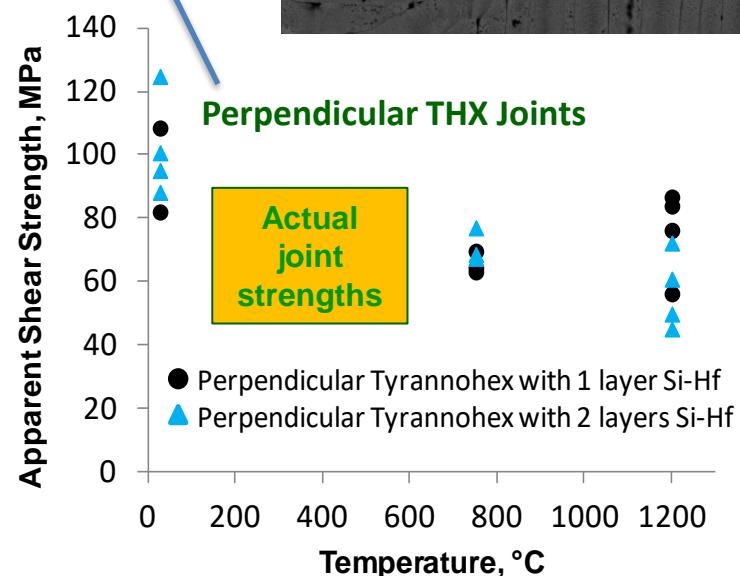
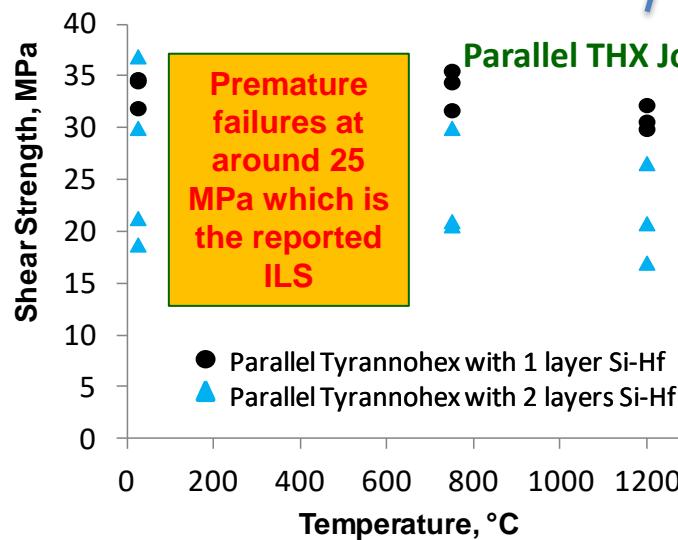
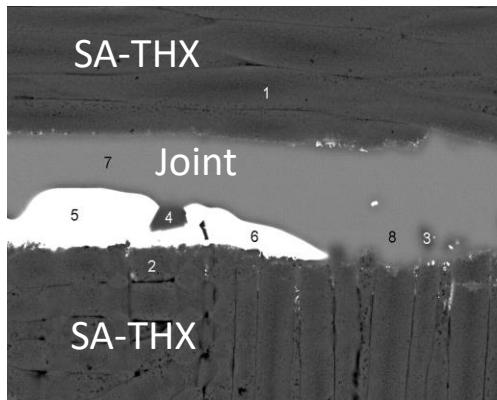
As bonded SA-THX in  $\parallel$  orientation (2 tapes)



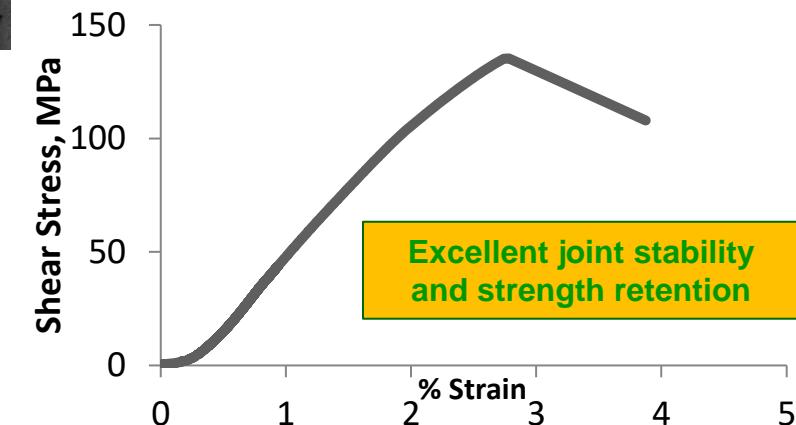
Joint region after SLO at 1200°C

Failure out of view in the composite

As bonded SA-THX in  $\perp$  orientation (1 tape)



Test configuration for single lap offset shear test



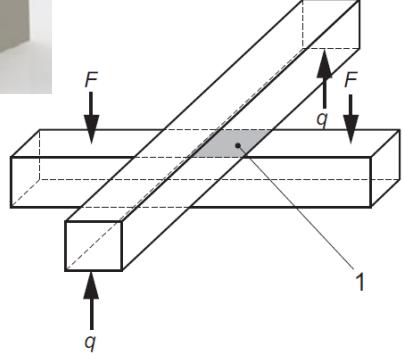
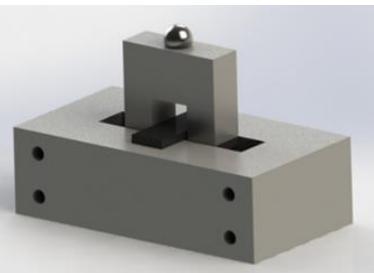
Residual Strength Test

- 350 hr run out at 1200°C and 25 MPa
- tested at 1200°C
- highest strength seen, 135 MPa

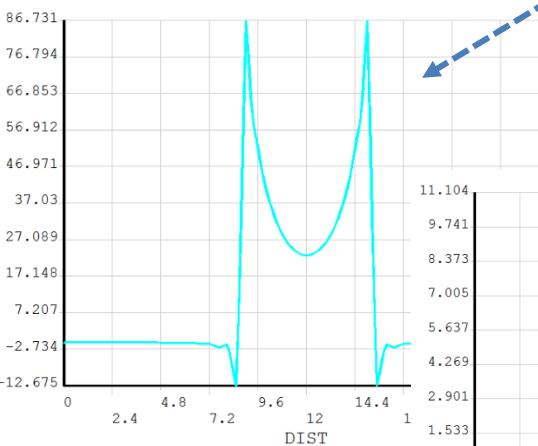
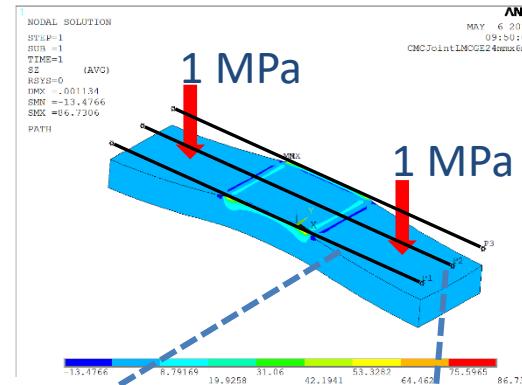
Composite interlaminar strengths and fiber architectures can cause premature failures.

Single lap offset shear test are good for in-house screening. REABond provides high shear and residual strengths.

# REABond Joining: Testing to ISO 13124



## Testing in Tension



**From FEA, Stress (MPa) versus location**



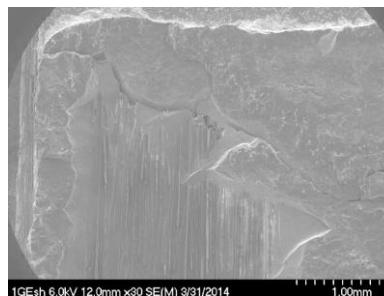
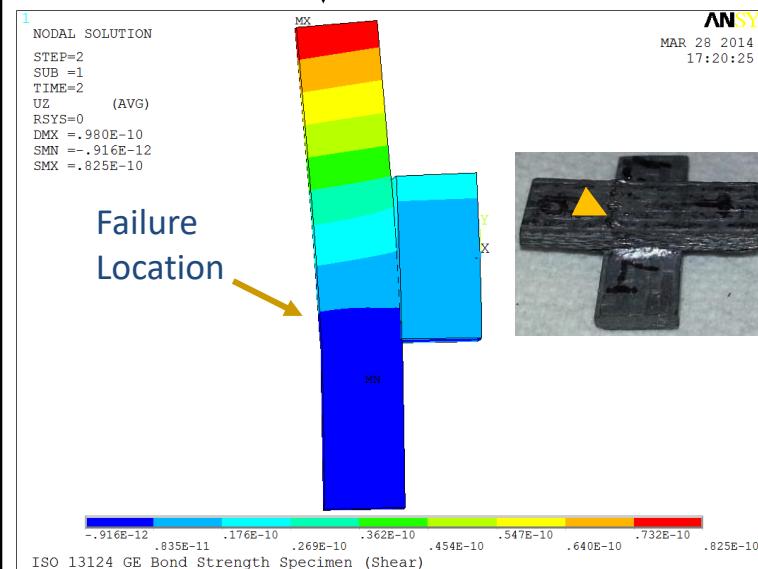
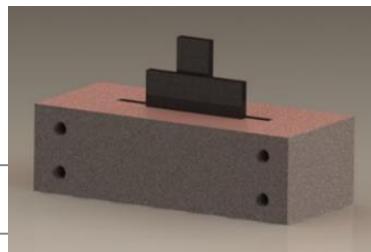
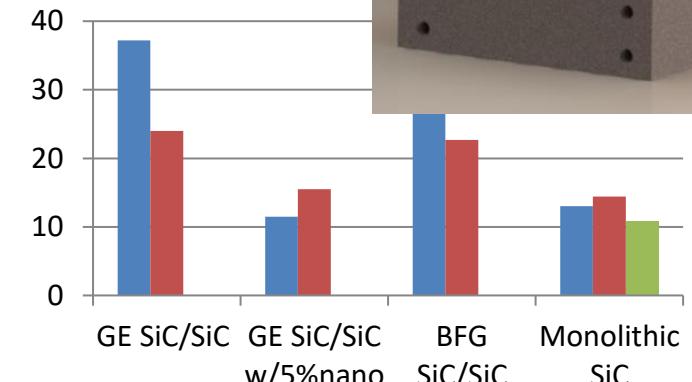
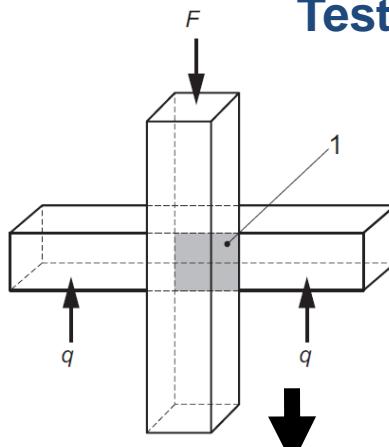
**BFG SiC/SiC Failure**



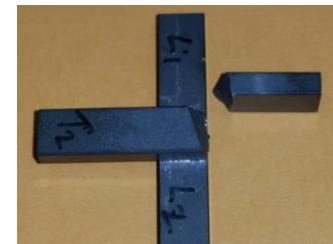
**SiC Failure**

M.C. Halbig, M. Singh, and J. Lang. "Development of High Temp. Joining and Thermomechanical Characterization Approaches for SiC/SiC Composites." *Advanced Processing and Manufacturing Technologies for Nanostructured and Multifunctional Materials II*, Vol. 36, Iss. 6 6 (2015): 3.

## Testing in Shear



**BFG SiC/SiC Failure**



**SiC failure**

The peak stresses, premature failures, low strengths, and fractures in the substrates demonstrate the challenges in having well developed and reliable test methods.

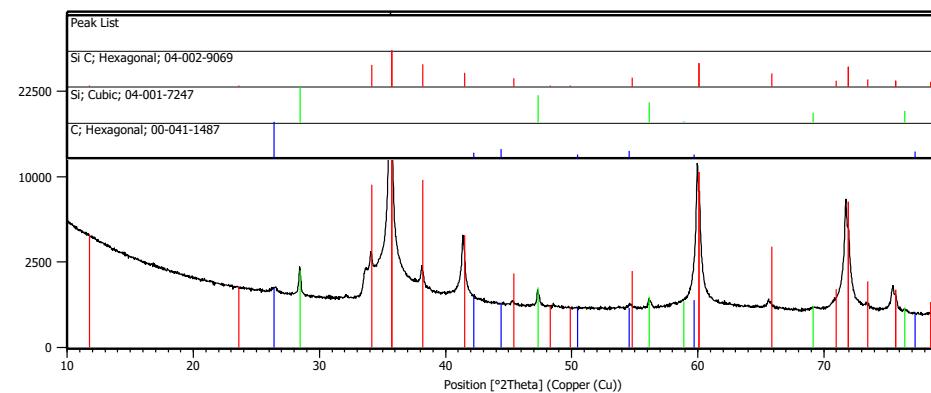
# SET: Single-Step Elevated Temperature Joining

## - Higher Temperature Capable C, Si, and SiC-Based Pastes

**Approach:** 30 mil thick green tapes of SiC, Si, and carbon powders of varying particle sizes, also several other additives.

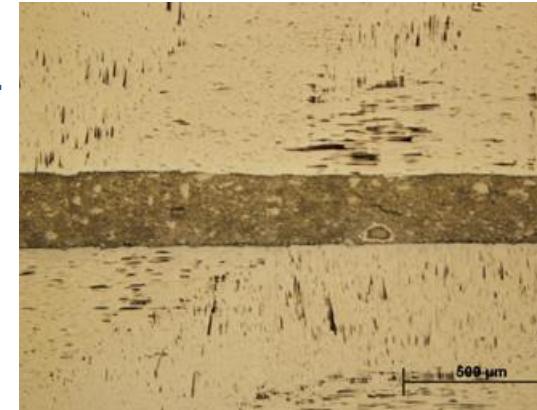
**Benefits:** high temp. capability and one-step SiC formation.

X-Ray Diffraction analysis of three slurry compositions heat treated at 1450°C for 30 min.

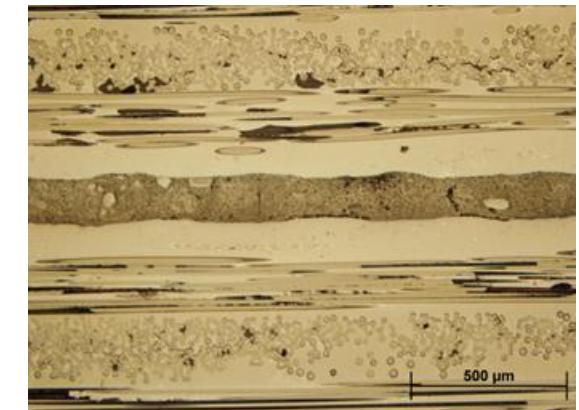


Composition	SemiQuant (%)		
	SiC	Si	C
J5A+Si	99	1	0 - nearly complete SiC conversion
J5A+N1+Si	91	9	1
J5A+N2+Si	92	7	1

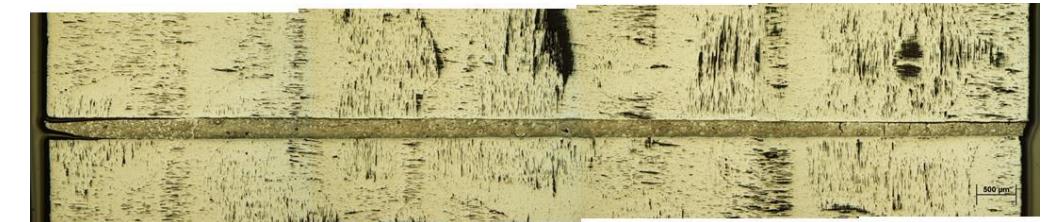
- High conversion to SiC provides one-step SiC formation.
- Offers low stress joining for complex shapes.
- High temperature capability (>2400°F) due to absence of free silicon.



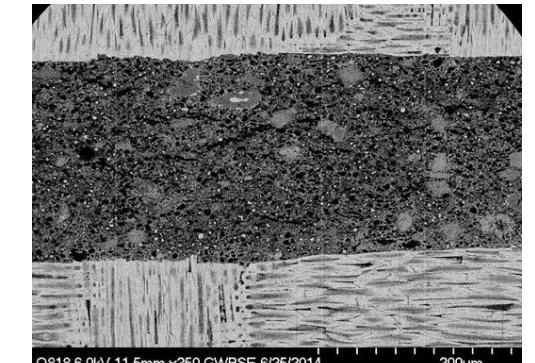
J5A+N2+Si Joining of SA-THX ( $\perp$ orientation)



J5A+N1+Si Joining of SiC/SiC



Perpendicular  
SA-Tyrannohex  
with N1+J5A+Si





## Summary/Conclusions

- Good quality joints are obtained from all five CMC to CMC joining methods: Brazing, ARCJoinT, Diffusion Bonding, REABond, and SET.
- Diffusion Bonding is the most restrictive joining approach requiring flat shapes, relatively smooth surfaces and high pressures.
- Brazing, REABond and SET approaches are the most versatile allowing for tailored interlayers for pressureless joining of complex shapes with smooth or rough surfaces in one-step processing.
- Particulate additions to the braze were shown to modify the hardness and thermal expansion of the joint.
- Mechanical tests to include ISO 13124 and single-lap offset shear are being used but additional analysis and improved test methods are needed.
- Selection of proper joining technologies is critical for the successful development and applications of ceramic components in a wide variety of current and emerging applications.
- These approaches offer many opportunities for development and implementation of turbine engine and other aerospace components with improved performance, lower emissions, lighter weight, and thermal management.

## Acknowledgements



- This work was support by the NASA aeronautics projects of:
  - Transformative Tools and Technology (TTT)
  - Revolutionary Vertical Lift Technology (RVLT, previously Subsonic Rotary Wing)
  - Hypersonics Technology Project (HTP).
- Thank you to John Setlock at NASA GRC for preparing REABond tapes.
- Thank you to Ron Phillips (retired) for ISO 13124 testing.
- Thank you to all collaborators/co-authors as noted in the referenced papers.

